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A Study of Nucleation and Growth of Thin Films by Means of Computer Simulation— General Features

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A STUDY OF NUCLEATION AND GROWTH OF THIN FILMS BY MEANS
OF COMPUTER SIMULATION - GENERAL FEATURES

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ABSTRACT

Some of the processes involved in the nucleation and growth of thin films were simulated by means of a digital computer. The simulation results were used to study the nucleation and growth kinetics resulting from the various processes. Kinetic results obtained for impingement, surface migration, impingement combined with surface migration, and impingement combined with surface migration and with reevaporation are presented. A substantial fraction of the clusters may form directly upon impingement. Surface migration results in a decrease in cluster density, and reevaporation of atoms from the surface causes a further reduction in cluster density.

INTRODUCTION

The kinetics of nucleation and growth of thin films has been the subject of experimental and theoretical study for a long time. The standard theoretical treatment is based on the rate equations derived by Zinsmeister¹ and modified by others²⁻⁶ to take into account the various processes involved, e.g., impingement, surface migration, reevaporation, and cluster formation. These equations cannot, however, be considered a faithful description of the process, since, whereas they are deterministic in nature and yield a single solution, the various processes involved in nucleation and growth are clearly random and cannot be expected to yield a repeatable result.

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The study reported here is an attempt of a different approach to the problem which takes into account this random nature of the processes involved, namely, computer simulation. Similar techniques have been recently applied to the study of aggregation and diffusion-controlled cluster formation⁷⁻⁹. In this paper only the general features of the results will be presented. A more detailed account of the results will be given elsewhere.

DESCRIPTION OF THE MODEL

A square lattice was selected for the simulation, since by far most of the experimental studies of nucleation and growth used as a substrate the (100) surface of NaCl, which is of this type¹⁰⁻¹⁶. The basic assumption made in the model are

- (1) Only single atoms impinge on the substrate surface.
- (2) Only single atoms are mobile on the surface.
- (3) Mobile atoms migrate on the surface by jumps to nearest-neighbor sites, and the jump frequency is constant.
- (4) Only single atoms can reevaporate from the surface.
- (5) Only nearest-neighbor interactions are taken into account.
- (6) A cluster is formed by two or more atoms occupying adjacent lattice sites.
- (7) No decomposition of clusters takes place.
- (8) The surface is free from sites that cause preferential nucleation, e.g., defects or impurities.
- (9) The simulation is limited to a single monolayer, i.e., to the early stages of the Frank-van der Merwe and Stranski-Krastanov mechanisms of film growth.
- (10) The boundary conditions taken are of an infinite potential barrier, i.e., atoms cannot cross the boundary.

Most of these assumptions were given and discussed by Logan². No claim is made as to the validity of any of them. (A study of the effect of at least some of them upon nucleation and growth behavior is planned.)

PROCEDURE

The simulations were carried out on a minicomputer. A lattice of 400 (20x20) points was the sample lattice. To check the effect of lattice size on the results, some test runs were also performed on a 1600-point (40x40) lattice. The results in these runs fell within the error range of the results obtained for the 400-point lattice.

The x and y coordinates of the sites of atom impingement were created by a random number generator with homogeneous distribution. A single atom on the surface can jump in one of the four directions shown in Fig. 1. The actual direction of each jump was also determined by means of a random number generator, which was programmed to yield as an output one of the numbers 1, 2, 3, or 4. Once two atoms occupied nearest-neighbor positions on the lattice they were not allowed to change their positions.

For each case studied eight simulation runs were performed, and the average as well as the standard deviation of the various parameters was evaluated. To check the sufficiency of the number of runs, 16 simulation runs were performed for some cases. The results were in the error range obtained with eight runs, and no reduction of the standard deviation resulted.

RESULTS AND DISCUSSION

To study the characteristics of each of the processes involved in nucleation and growth, some of the simulations were first done separately, and then the combined process was studied. The simulations were used to estimate the number of atoms nucleated (i.e., atoms in clusters), the cluster density, and the size distributions of clusters.

Impingement

Some of the clusters are formed directly on impingement without any surface migration. The physical process simulated in this case is the one that takes place when a film is deposited on a very cold substrate. The results for the number of atoms nucleated as a function of time are presented in Fig. 2. A substantial fraction of the impinging atoms are trapped in clusters, this fraction increases as the process proceeds. This is demonstrated better in Fig. 3 where the percentage of nucleated atoms out of the total number of impinging atoms is plotted versus time. Figure 4 shows the cluster density resulting from direct impingement. The density first increases, reaches a maximum at a coverage of ≈ 0.4 , and then decreases due to the overlap of clusters.

Surface Migration

Next, the effect of surface migration following impingement was studied. Physically, this would correspond to the case where a film is deposited on a very cold substrate which is later warmed up to allow migration of atoms on the surface. The results reported here (Figs. 5 and 6) are only for one value of coverage, $\theta = 0.1$. The process reaches saturation after 87 ± 24 jumps. Comparison of Fig. 6 with Fig. 4 allows one to estimate the fraction of clusters that are formed directly on impingement relative to the total number of clusters obtained after migration is completed. The number of clusters per site formed on impingement at this coverage is 0.013 ± 0.002 , the number of clusters per site following migration is 0.034 ± 0.003 . Thus, some 28 ± 5 percent of the clusters are formed directly on impingement, and the remaining 72 ± 5 percent are due to surface migration.

Combined Impingement and Migration

This case of combined impingement and migration corresponds to the situation where a film is deposited on a substrate that is hot enough to allow surface migration yet is too cold to allow reevaporation of adsorbed atoms. The simulation results, for the case where the impingement rate is 2.5×10^{-3} atom/(site)(unit time) and the jump frequency is 1 per unit time, are presented in Figs. 7 to 9. The effect of surface migration on growth kinetics is best seen by comparing Fig. 3 with Fig. 8, where the percentage of nucleated atoms out of total number of impinging atoms is plotted versus time.

The effect of surface migration on nucleation kinetics, i.e., on the cluster density, is revealed by comparing Fig. 4 with Fig. 9: Surface migration causes a substantial decrease in cluster density.

Reevaporation

Finally, the effect of the reevaporation of adsorbed atoms from the surface on the nucleation and growth behavior was studied. Only one case is presented here, namely, the one where the probability of each single atom reevaporating is 0.5. In other words, on each jump every single atom has an equal chance of either landing on an adjacent lattice site or leaving the surface.

The growth rate results for this case are given in Fig. 10. It is worth noting that in this case almost all the atoms on the surface at any given time are bound in clusters, since the ones that are not have little chance of surviving there. This has the effect of about a fourfold slowing of the growth kinetics, as can be seen by comparing Fig. 10 with Fig. 7. The effect on nucleation kinetics is shown in Fig. 11 where a threefold reduction in cluster density, compared with the case of no reevaporation (Fig. 4), can be seen.

CONCLUSIONS

1. Because of the random nature of the various processes involved in nucleation and growth, a large scatter in results is obtained. This is reflected in the high values of the standard deviation.
2. Depending on impingement rate and jump frequency, a substantial fraction of clusters may be formed directly upon impingement.
3. Surface migration results in a decrease in the cluster density.
4. Reevaporation of atoms from the surface causes a further reduction in cluster density.

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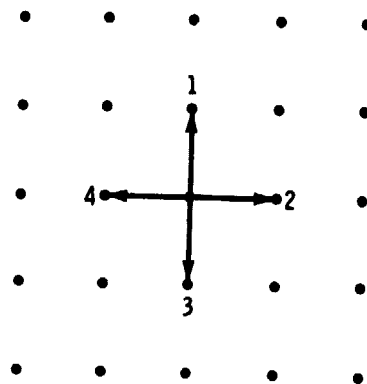


FIG. 1. Designation of jump directions.

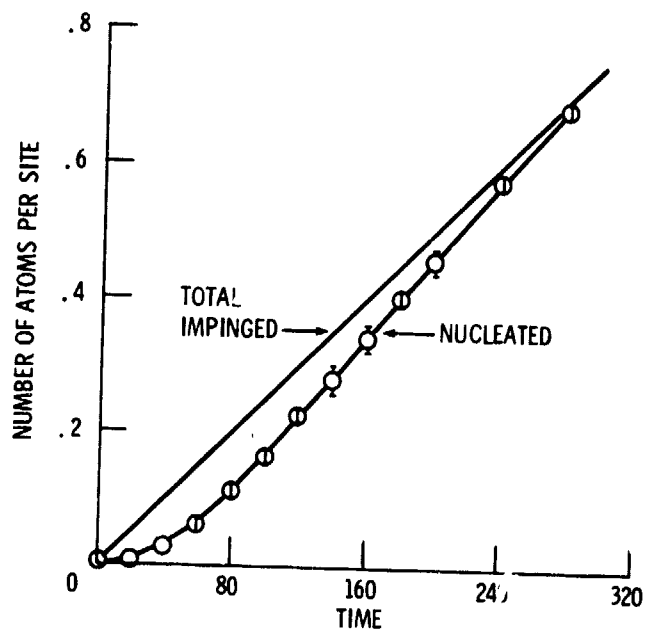


FIG. 2. Atoms nucleated directly upon impingement.
Impingement rate, 2.5×10^{-3} atom/(site x unit time).

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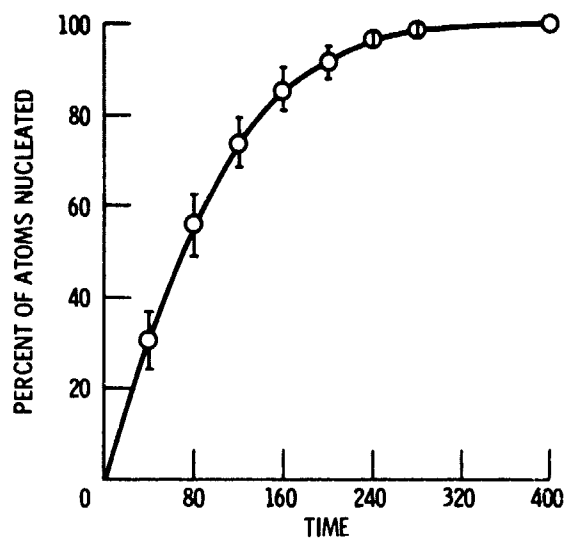


FIG. 3. Percent of atoms nucleated directly upon impingement. Impingement rate, 2.5×10^{-3} atom/(site x unit time).

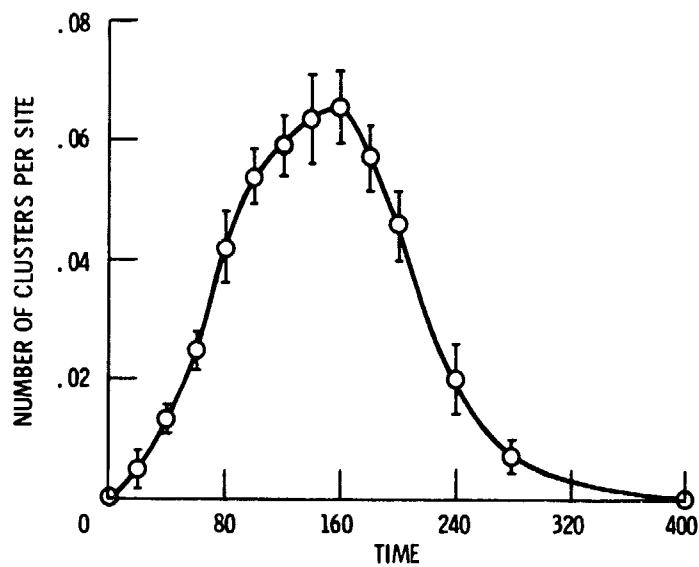


FIG. 4. Cluster density due to impingement. Impingement rate, 2.5×10^{-3} atom/(site x unit time).

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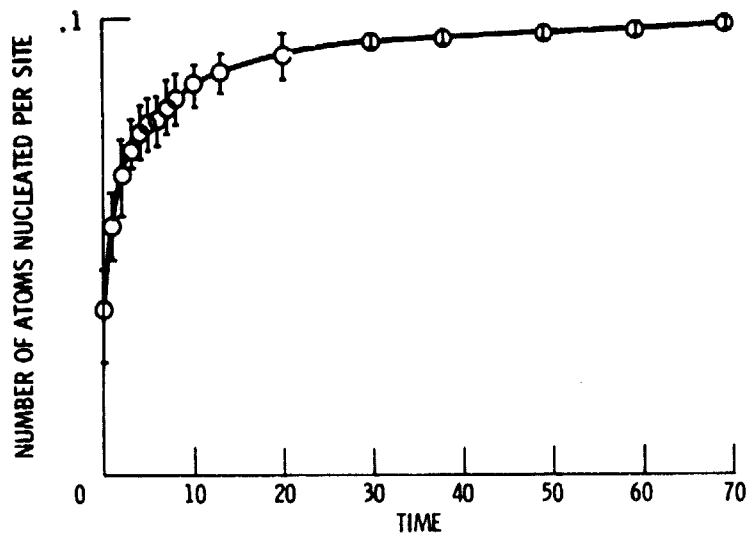


FIG. 5. Nucleated atoms due to surface migration. No impingement; jump frequency, 1 per unit time; coverage, 0.1.

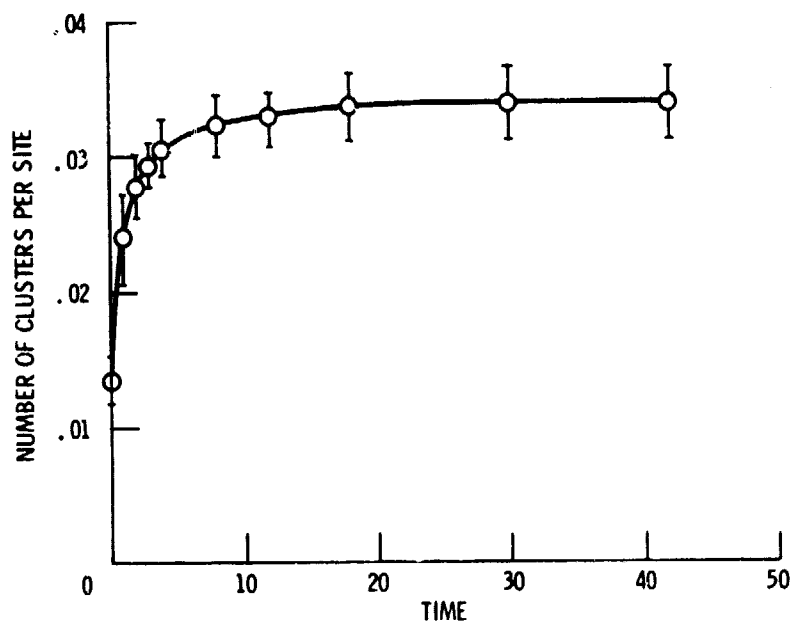


FIG. 6. Cluster density due to surface migration. Jump frequency, 1 per unit time; coverage, 0.1.

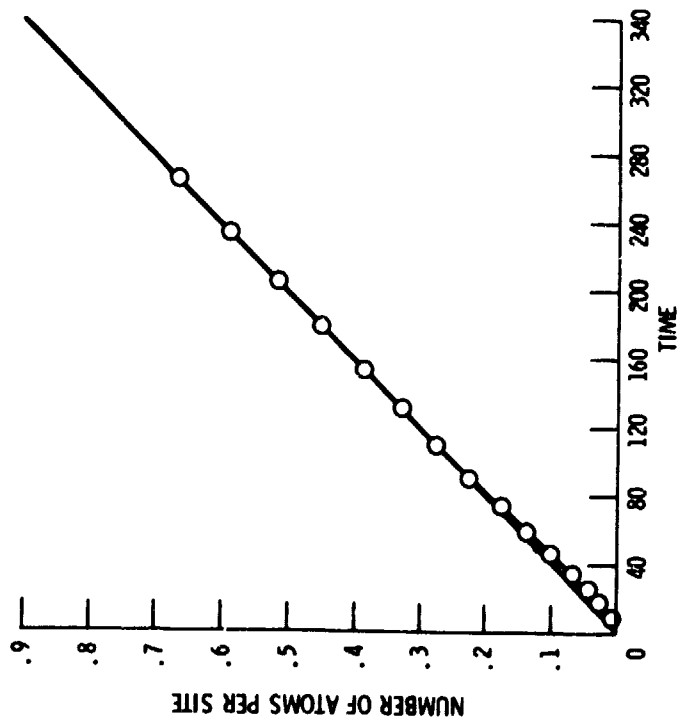


FIG. 7. Nucleated atoms due to combined impingement and surface migration. Impingement rate, 2.5×10^{-3} atom/(site x unit time); jump frequency, 1 per unit time.

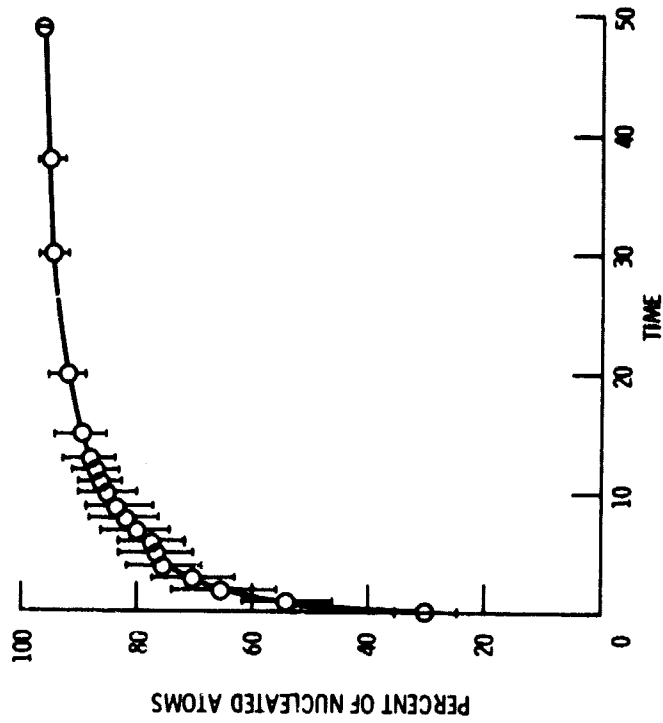


FIG. 8. Percent of nucleated atoms on combined impingement and surface migration. Impingement rate, 2.5×10^{-3} atom/(site x unit time); jump frequency, 1 per unit time.

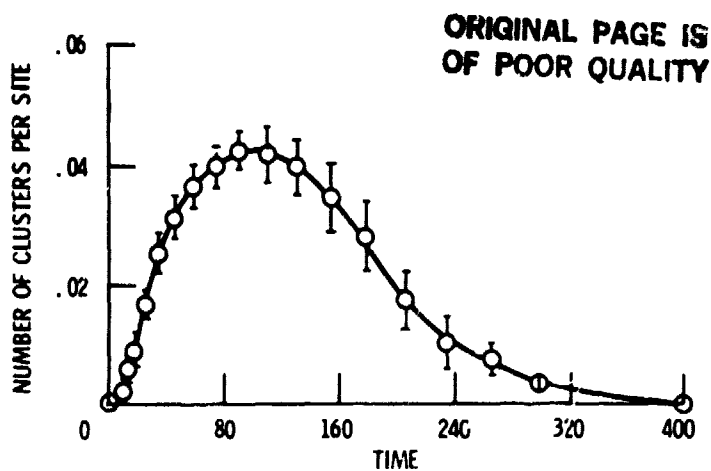


FIG. 9. Cluster density due to combined impingement and surface migration. Impingement rate, 2.5×10^{-3} atom/(site x unit time); jump frequency, 1 per unit time.

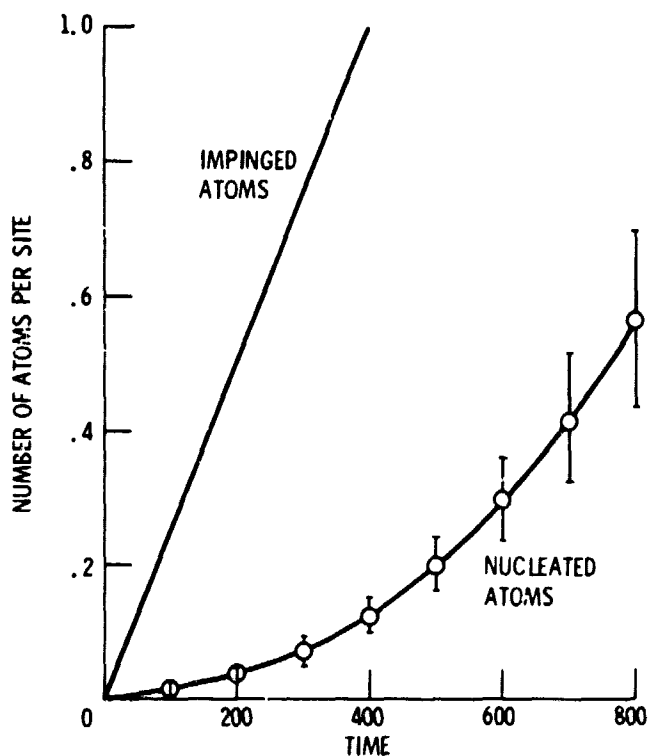


FIG. 10 Nucleated atoms due to combined impingement, surface migration, and reevaporation. Impingement rate, 2.5×10^{-3} atom/(site x unit time); jump frequency, 1 per unit time; coverage, 0.5.

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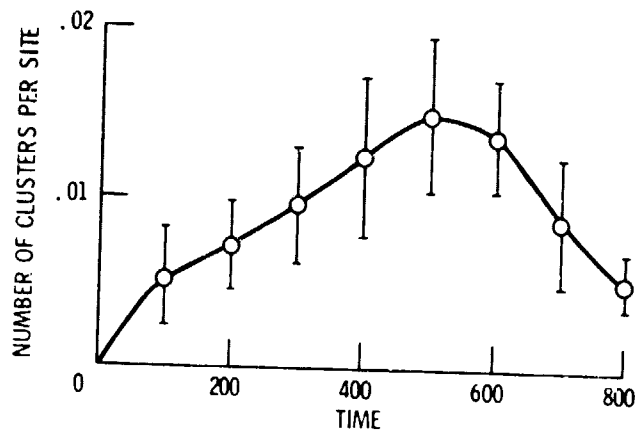


FIG. 11. Cluster density due to combined impingement, surface migration, and reevaporation. Impingement rate, 2.5×10^{-3} atom/(site x unit time); jump frequency, 1 per unit time; reevaporation probability, 0.5.